

Citation for published version:

Blaney, A, Dunn, NS, Alexander, JM, Richards, D, Rennie, AEW & Anwar, J 2017, 'Directing self-assembly to grow adaptive physical structures', *International Journal of Rapid Manufacturing*, vol. 6, no. 2-3, pp. 114-133. <https://doi.org/10.1504/IJRAPIDM.2017.10003069>

DOI:

[10.1504/IJRAPIDM.2017.10003069](https://doi.org/10.1504/IJRAPIDM.2017.10003069)

Publication date:

2017

Document Version

Publisher's PDF, also known as Version of record

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Directing self-assembly to grow adaptive physical structures

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Abstract: Additive manufacturing technologies offer exciting opportunities to rethink the process of designing and fabricating physical structures. This paper outlines initial work that seeks to extend existing AM capabilities, creating physically adaptive structures by exploiting processes of self-assembling materials. The paper details an investigation of self-assembling structures that can respond to different conditions by adapting their physical properties over time. The process uses electrolysis of seawater to demonstrate a proof-of-concept of tuneable material structures, via crystal growth. Results demonstrate an aggregation-based multi-material system that is sensitive to changing environmental conditions. Material properties of grown structures have been analysed and illustrate that different materials can be created from an abundant base material (seawater) by manipulating environmental conditions (i.e. electrical current). It is found that turbulence is a useful property within these kinds of systems and that the physical properties of cathode scaffold structures have a significant impact in controlling material properties and resolution.

Keywords: adaptive materials; additive manufacturing; self-assembly.

Reference to this paper should be made as follows: Blaney, A., Alexander, J., Dunn, N., Richards, D., Rennie, A. and Anwar, J. (2017) 'Directing self-assembly to grow adaptive physical structures', *Int. J. Rapid Manufacturing*, Vol. 6, Nos. 2/3, pp.114–133.

Biographical notes: Adam Blaney is a PhD candidate based at the Highwire CDT at Lancaster University. Graduated from the Manchester School of Architecture (MSA) in 2010 (BA) and 2012 (BArch). His research focuses on developing a discourse between design, fabrication and material properties to create adaptive, tunable physical structures.

Dr Jason Alexander is a Senior Lecturer in the School of Computing and Communications at Lancaster University. His primary research focus is in Human-Computer Interaction, with a particular interest in developing novel interactive systems to bridge the physical-digital divide. His recent work focuses on the development of shape-changing interfaces-surfaces that can dynamically change their geometry based on digital content or user input. Alexander's other research has investigated novel design processes for personal fabrication, and new interaction techniques using eye-gaze, haptic feedback and gestural interaction.

Professor Nick Dunn is Executive Director of Imagination Lancaster, an open and exploratory design research lab at Lancaster University, where he is also Chair of Urban Design. He is Associate Director of the Institute for Social Futures, where he also leads research in the Future of Cities and Urbanism. His work responds to the contemporary city as a series of systems, flows and processes and is explored through experimentation and discourse addressing the nature of urban space: its perception, demarcation and appropriation. In particular, he is interested in why and how (maybe even where and when) we design, rather than what we design. He has published numerous books related to architecture, digital fabrication and urbanism and his papers have been published and presented internationally, with collaborative creative work exhibited across the UK, China and the Ukraine.

Dr Daniel Richards is a Lecturer in Data Prototyping and Visualisation at Lancaster University. After studying architecture, his PhD (Manchester Metropolitan University) explored automatic synthesis of architectural designs with evo-devo inspired algorithms. He subsequently undertook postdoctoral research in the Informatics Research Centre at Manchester Metropolitan

University focusing on evolutionary design of engineering structures. His ongoing research combines engineering design, 3-D modelling and visualisation, nature-inspired computation and digital fabrication to explore the future of manufacturing.

Dr Allan Rennie graduated from Glasgow Caledonian University in 1995 with a BSc in Computer Aided Engineering, before moving to the Centre for Rapid Design and Manufacture at Buckinghamshire College and graduating with a PhD in additive manufacturing from Brunel University in 2001. In 2002, Allan moved to Lancaster University and established the Lancaster Product Development Unit (LPDU), using EU funding to enable the development of collaborative research relationships between academia and the private sector with a focus towards engineering design, product development and additive manufacturing. To date, LPDU have successfully delivered projects in excess of £13M, collaborating with in excess of 750 businesses and other organisations. Whilst retaining his function as Director of LPDU, in 2012 Allan accepted an academic role and is also currently a Senior Lecturer in Manufacturing Engineering within the Engineering Department at Lancaster University.

Professor Jamshed Anwar is an Associate Dean for Research for the Faculty of Science & Technology at Lancaster University, and holds a Chair in Computational Chemistry. His research interest is phase transition phenomena involving molecular assemblies which he studies using computational modelling and simulation. A particular interest is crystal nucleation and growth and associated phase transitions. Prior to his appointment at Lancaster, he has held posts at the University of Bradford and at King's College London. He has also spent periods at the University of Pennsylvania, the Institute of Atomic & Molecular Physics (AMOLF) in the Netherlands, and the University Chemical Laboratory, Cambridge. He was awarded the Pfizer Award in 1999 for his seminal studies on molecular simulations of pharmaceutical systems, and is also a recipient of the RP Scherer Award (1986). He holds visiting professorships at a number of leading institutions including the Chinese Academy of Sciences Institute of Materia Medica, Shanghai, China.

This paper is a revised and expanded version of a paper entitled 'Adaptive materials: utilising additive manufactured scaffolds to control self-organising material aggregation', presented at *14th Rapid Design, Prototyping and Manufacturing Conference*, 15–16th December, 2015, Loughborough Design School, Loughborough University.

1 Introduction

Biological structures continually adapt their shape and material compositions to suit environmental demands, maximise available resources and have the ability to heal. These properties are possible because biological materials are not inert. That is, in contrast to traditional manufactured materials, biological materials can adjust their properties 'on-the-fly' and self-organise. For example, bone modelling and remodelling (AMGEN, 2012) is a process that *tunes* the mechanical and material properties (density/strength) of bone over time to address regular physical stresses acting on them. Biological fabrication processes use external forces to inform material deposition (Vogel, 2003), producing extremely multifunctional and materially economic structures.

Manufactured structures, which adapt their physical properties to meet fluctuating demands, could offer exciting new design possibilities. For example, building components that could alter their material properties (e.g. porosity, elasticity, rigidity, density) to meet temporal demands so as to avoid structural failure when new physical loads are introduced; or machine parts that heal themselves when damaged. Adaptive synthetic materials would also open up new possibilities in the fashion industry, such as wearable materials which change colour or pattern on demand. Finally, tuneable materials would allow radically new medical implants, prosthesis and splints to be designed, whereby the structures tune their prostheses (in real time) to meet unique patient physiology, thereby improving comfort or alternatively could be implanted as medical scaffolds to help regrow important biological material such as bone (Howes and Laughlin, 2012).

This paper will highlight the potential to instil adaptive qualities that are present in biological structures into manufactured ones by fabricating them out of self-assembling/self-organising materials. The key benefits of these adaptive materials/structures would be:

- 1 *Longevity* as changing and multiple demands can be accounted.
- 2 *Reduction of material waste* as resources can be redistributed within the structure/system over its lifetime.
- 3 *Scalable fabrication processes* that allows logic from digital design processes such as parametric design (Schumacher, 2016) to be embedded within them, which in turn may lead to more complex structures being created (Tibbits, 2012a, 2012b), which can physically *respond* and *adapt*.

An emerging area of architectural design is now exploring how advanced fabrication technologies can be combined with new materials to create adaptive, high-performance designs (Soldevila, Royo and Oxman, 2015; Oxman and Rosenberg, 2007; Tibbits and Cheung, 2012). This paper describes ongoing research towards this goal, demonstrating proof of concept physical structures that have tuneable adaptive material properties. The paper is structured as follows: first, a discussion of work relating to architectural design, synthetic biology and physical computing that uses adaptive and living materials to create structures; second, an outline of the experimental method that uses physical computing with crystal formation to build primitive tuneable materials; third, the initial results presented. Finally, a discussion of the findings and suggested opportunities for further work.

2 Background

Additive manufacturing (AM) enables new ways of constructing physical objects with novel material compositions (Oxman, 2011), so that they can fulfil multiple functions (Oxman, Keating and Tsai, 2011; Oxman et al., 2015) and even respond to external forces by changing shape in useful ways (Gutttag and Boyce, 2015). Interestingly, AM can also fabricate structures which act as growth media, such as agar. In this way, fabricated structures can be coupled with novel materials that grow within these media, allowing material properties to change over time within the defined forms (BioBot, 2016; Natalie Alima - *Bio Scaffold*, 2015). Another approach to create adaptive materials and

architectures utilises synthetic biology (Dade-Robertson, Ramirez and Zhang, 2014) and chemical systems (Armstrong and Spiller, 2010; Armstrong, 2014; Hanczyc, 2009). The former uses bacteria and the latter protocells, which can be programmed to respond to fluctuating environmental conditions, which then alters material properties.

A major challenge for these sorts of systems is how to direct growth processes and build useful structures. Note that in traditional CAD approaches, digital designs retain great flexibility whilst in the computer. The computer enables designers to change the shape of digital structures and or material properties in real-time. However when the digital design is physically fabricated using traditional CAM approaches, all of the flexible and adaptive behaviour within the digital model is lost. The designs become fixed and the physical objects do not adapt to changing conditions. In contrast, the huge promise of adaptive tuneable material is that they may enable *physical* objects to retain the same degree of flexibility, control and adaptability as their digital counterparts.

To explore new approaches to create adaptive materials for design and engineering applications, self-assembling materials have been incorporated into the fabrication process. Specifically, in this paper, the combination of self-assembling crystal materials with AM to achieve adaptive tuneable materials, mimicking bones' adaptive and tuneable abilities is proposed.

Mineral accretion (Hilbertz and Goreau, 1996; Hilbertz, 1992) is a process that deposits limestone (CaCO_3) and/or brucite ($\text{Mg}(\text{OH})_2$) on cathode structures that are submerged in seawater/brine using the process of electrolysis. Material build up on the cathode scaffold can be affected by multiple conditions, for example, distance from the anode, electrical current, cathode geometry and fluid flow (Goreau, 2012). Higher electrical voltages and current values create brucite quickly compared to lower voltages and current producing limestone slowly; altering these conditions over time can create a composite material (Corrie Van Sice Contributor, 2011).

Self-assembling materials (such as crystal growth) can adapt their physical properties (shape, density, composition, porosity, colour) in response to the physical environmental conditions in which they grow (temperature, pH, voltage). The objective of this research is to prove that by designing and manipulating cathode scaffolds, it is possible to direct how crystal structures grow. By carefully manipulating environmental conditions (e.g. electrical current, temperature, pH of the solution), it is possible to adapt and tune the deposited material properties (shape/location, volume, growth rate, composition, textures), and these studies may provide important clues for designing powerful AM technologies in the future that operate through directed self-assembly of materials.

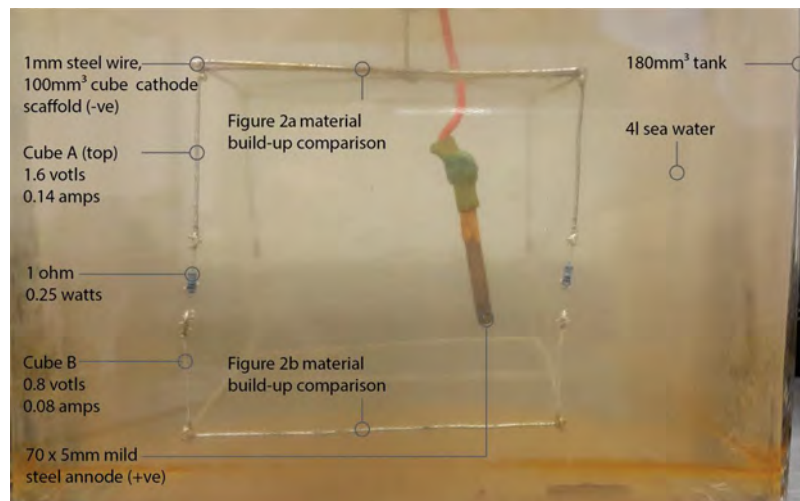
3 Methods

Currently, the proposed system uses the *mineral accretion* process to build adaptive structures that mimic adaptive and tuneable features of biological structures, such as bone. The mineral accretion process is relatively straightforward; a series of cathodes are placed within a seawater solution, and then by changing the electrical current, different types of materials (e.g. limestone and/or brucite) are deposited on the cathode scaffolds. The main benefit of this process is that it is robust, low cost (at the laboratory scale) and easy to setup initially. The current system controls material properties (location, volume, rate and type) manually by varying the voltage/current by connecting individual and or multiple cathode elements to be supplied with a fixed or varying voltage over time. This

creates a locally ‘tuneable’ environment that informs material aggregation, specifically, the location, volume, rate and composition of the material deposited on the scaffold.

Figure 1 details a basic cathode setup. The cathode incorporates four resistors, which is intended to allow different values of electrical current to be present at different parts of the cuboid cathode structure. The cube cathode was submerged in natural seawater. The varied electrical current values created across the cube cathode because of the resistors and the composition of seawater solution are used to control where local material is deposited and its properties (i.e. mix of limestone and brutice).

Figure 1 Cuboid cathode with resistors to create two different currents in this configuration. The first experiment was carried out for 240 min and material deposition recorded every 30 min. A benchtop power supply unit was used to supply electrical current (see online version for colours)



The paper now presents two experiments, which use different cathode designs to control material aggregation in specific ways.

4 Results

To test the model, two experiments were performed. The first experiment provides a proof of concept that the model can indeed generate multi-material structures through the mineral accretion process, whilst the second experiment shows that material properties can be tuned to create different shapes and material compositions by adjusting controllable parameter such as voltage.

4.1 Experiment 1: proof of concept material aggregation

To test that the model is able to generate multi-material structures through the mineral accretion process, the cathode cuboid shown in Figure 1 was submerged in seawater and 1.6 V and 0.14 A were applied to the top elements of the cube, the resistors then reducing

the voltage to 0.8 V and 0.08 A to the bottom elements. The system was operational for 24 h.

It was predicted that more material could accumulate on the top of the cube, and the material would be predominantly brucite as it was supplied with an increased voltage and current, which results in the growth of brucite, which grows faster than limestone [2222]. Similarly, it was expected that the bottom elements of the cube experience less material aggregation, yet the material to be predominantly limestone.

To perform material analysis on the results, a scanning electron microscope (SEM) was used to reveal different material types. Under SEM, brucite resembles rosette formations, whereas limestone is shown as needle-like shapes [22]. Additionally, fine powder samples from the aggregated materials were analysed using X-Ray diffraction (XRD), which provided a quantitative measure of the material properties.

Examination of the material deposited on the cuboid cathode revealed that most material accumulated at the bottom (Figure 2b), rather than the top elements as expected. XRD analysis revealed that brucite was more predominant on the top elements of the cube than at the bottom of the cathode structure, as expected, meaning that the different currents did appear to have some effect, yet the overall aggregation process was highly susceptible to the effects of gravity, leading to much more material settling on the bottom of the cube compared to the top parts.

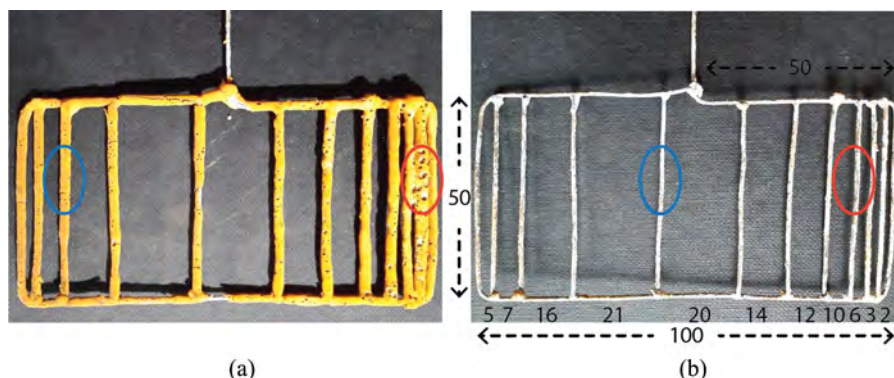
Figure 2 (a) Cuboid material build-up top. Top of cube has less material than the bottom. (b) Cube material build-up bottom. More material accumulated at the bottom of the cube due to gravity. The wire is 1 mm in diameter (see online version for colours)



To improve on these results, three key changes were made to the experimental setup. First, improving the consistency of the seawater solution by dissolving 100 g of marine salts in 3 L of water at 25°C. Second, adding an aquarium wave maker to the system to provide agitation of the solution at a rate of 2,000 L/h. Finally, changing the shape of the cathode setup, testing two further options:

- 1 where all elements lay on the same plane and resembled a flat fence structure with irregular spacing between vertical elements (Figure 3)
- 2 with two individual straight cathode wires which were hung vertically in the seawater tank.

Figure 3 (a) Cathode shape informs material properties deposited. The substance on Figure 4a is created from the dissolved anode (iron oxide). (b) Initially a uniform material comprised of limestone and brucite was deposited but then inhibited as the dissolved anode material predominated. All units in mm. 1 mm diameter steel wire used to make the cathodes (see online version for colours)

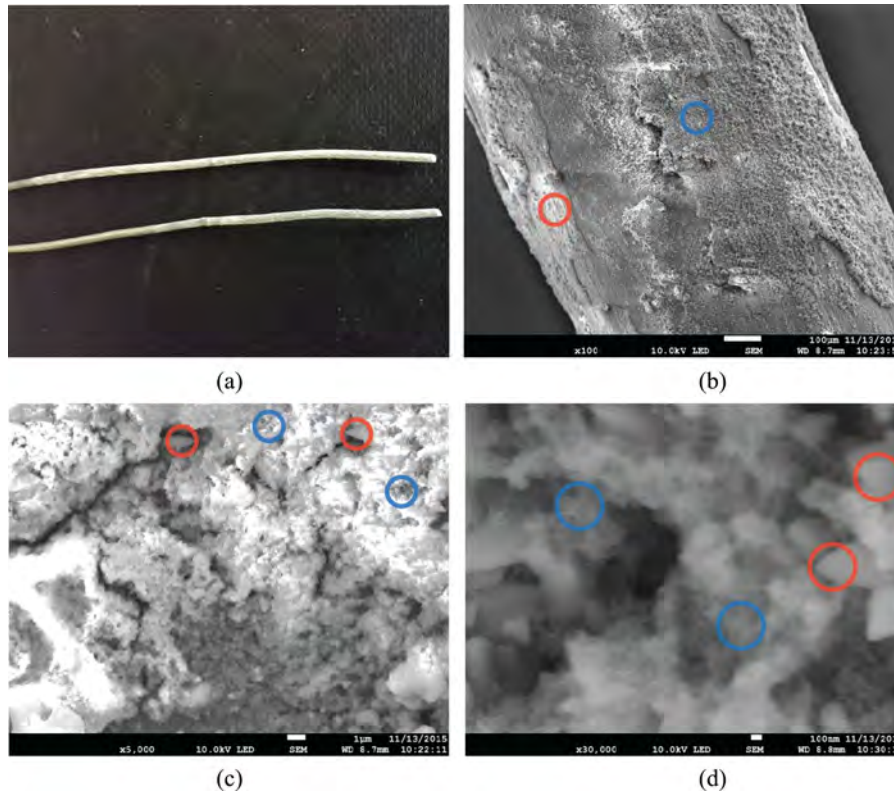


The system was once again run for 24 h, and SEM and XRD material analyses were undertaken.

Figure 3 shows the fence cathode with two material build-ups occurring. Material substance shown on Figure 3a is a result of the steel anode dissolving, which contaminated the solution. As a result, this material growth predominated after the initial white material. XRD analysis of the porous material substance reveals that it is a foreign material as it does not match either the known calcium carbonate or magnesium hydroxide values. Examination of the material's surface in Figure 3a revealed a porous quality, which increased as the vertical elements of the fence became closer to one another. The material also began to join together between the closer vertical elements, resulting in a solid wall-type build-up. The material shown in Figure 3b was initially grown on the fence cathode, as it was revealed when the porous material was removed. Notably, the XRD analysis did reveal that both limestone and brucite were present in the material samples, with limestone being more predominant, which suggests that a multi-material structure could indeed be built with some modification to the cathode structure.

Figure 4 shows the two individual straight cathode wires which were analysed by way of SEM. The SEM analysis from a portion of a wire reveals varying amounts of limestone and brucite, which was dependant on the location where the material analysis was undertaken. Interestingly, this revealed that the purity of the cathode material also affects material type deposited, because it influences the electrical current uniformity. This finding suggests that altering the current value within the cathode scaffold controls the material type deposited. However, the results from the cuboid cathode experiment also suggest that agitating the solution is the overriding factor to control material location, rate and volume deposited. Examination of the fence cathode found that agitation counteracted the effects of gravity and created a more uniform material deposition (Figure 3).

Figure 4 (a) Cathode wire to test material build-up using SEM. (b) SEM revealed effects of the cathodes purity as varying amounts of brucite and limestone crystals depending on analysis locations. (c) and (d) Brucite highlighted with red circles compared to the limestone highlighted with blue circles. 100 × 1-mm-diameter steel wire cathode used (see online version for colours)



Finally, Figures 5 and 6 present the data from these experiments. Figure 5 shows the material composition by way of SEM analysis of the two straight wire cathodes, which reveals the varying amounts of limestone and brucite build-up that occurred in two different locations. The varying material composition was a result of the non-uniform material properties of the wires which created varying electrical currents over its surface. Figure 6 then compares XRD values of the material build-up taken from the cube and fence cathode to known values for limestone and brucite. These results establish proof of concept that it is possible to govern material properties of the mineral accretion process. This was achieved by physically manipulating the environment in which the materials grow (voltage and agitation) as well as alter the cathode scaffolds' properties on which the material was deposited.

Figure 5 SEM analysis of two wire cathodes reveals varying amounts of brucite and limestone depending on analysis location. This establishes the effects of the cathodes' purity (how uniform the electrical current is) on material type. This establishes a multi-material system is achievable using this process by fluctuating current (see online version for colours)

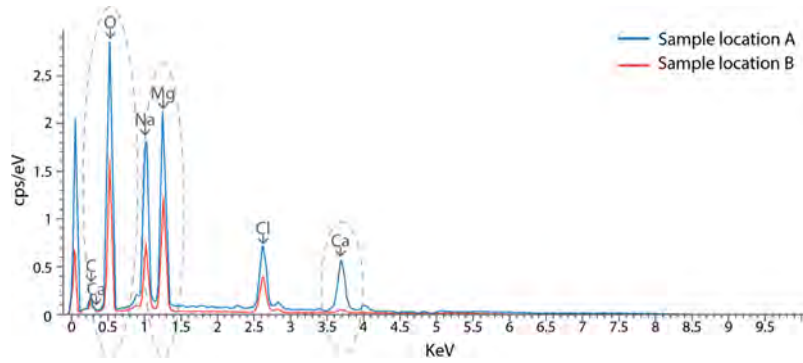
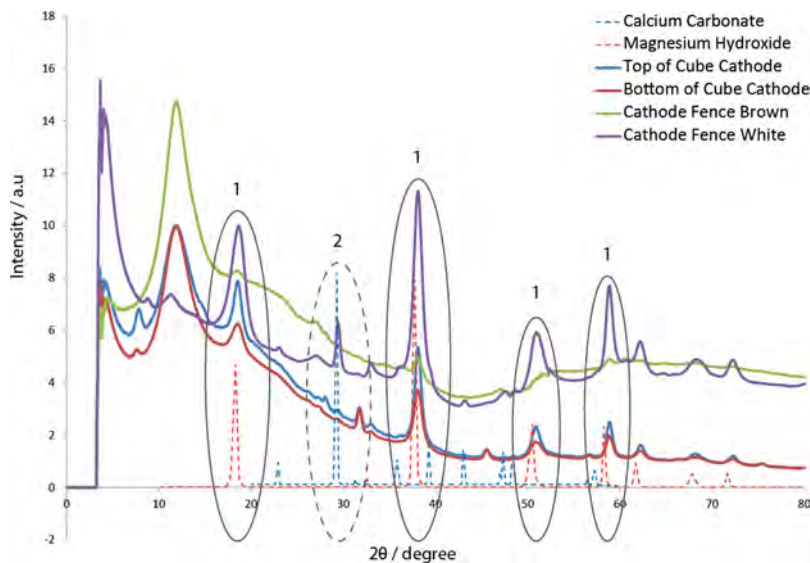


Figure 6 XRD of samples compared to CaCO_3 and $\text{Mg}(\text{OH})_2$. The cube cathode strongly resembles $\text{Mg}(\text{OH})_2$ values (circles with 1), demonstrating that the cube cathode typology allows limited control. Whereas analysis of the white fence cathode demonstrates that both material types are created (circles with 1 and 2), and therefore by controlling electrical current, it is possible to tune material properties within a range (see online version for colours)



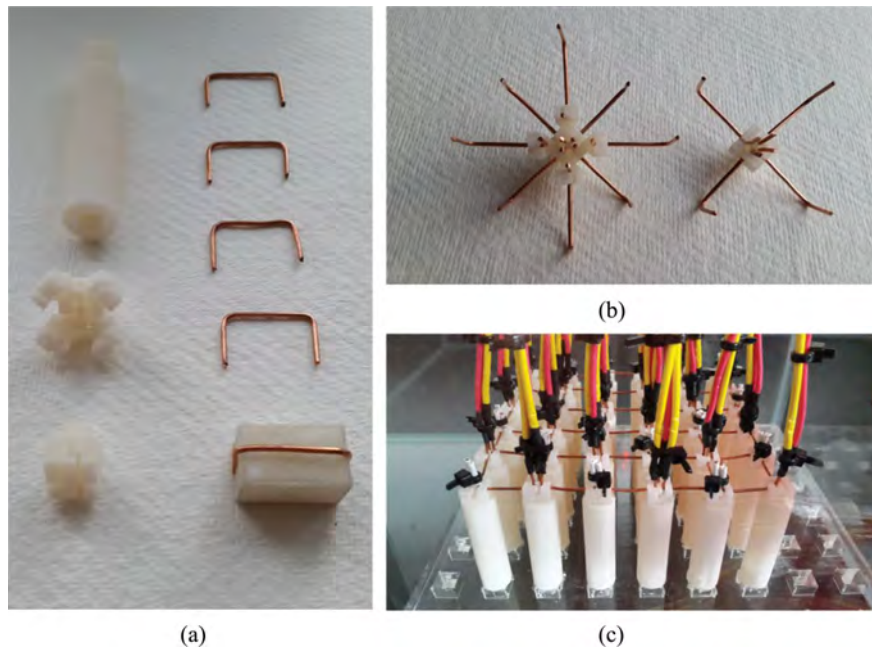
4.2 Experiment 2: tuneable materials on a 2D grid

To extend the proof of concept study and demonstrate that simple shapes and material composition can be controlled through material aggregation and directed self-assembly, a 6×6 2D grid of conductive elements (Figure 2) was used to grow a specific shape with varying material thicknesses. The effects of varying voltages supplied to individual

elements on the grid will determine its effect on material composition at defined locations.

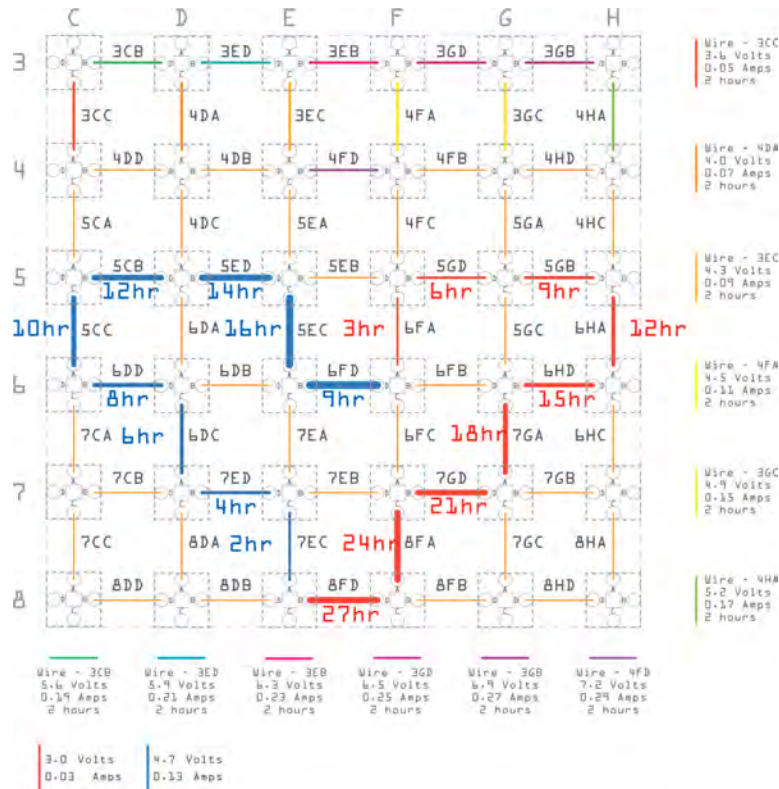
Figure 7 describes the 6×6 2D cathode grid, which is made up of physically separated cathode elements (1-mm copper wires). Material build-up on the grid cathode is manually controlled by connecting individual elements or multiple elements (through a breadboard) to a bench power supply. The conductive copper wires (i.e. the cathode) are held in place using modular components, which were fabricated in insulating nylon with AM (using the selective laser sintering process). The modular supports are used to physically separate the copper wires and allow specific electric currents to be applied across the 6×6 -gridded cathode structure.

Figure 7 (a) AM was used to fabricate modular nodes made to physically separate the cathode elements. (b) Two different heads were fabricated to increase the resolutions of the grid. (c) Final 2D 6×6 grid (see online version for colours)



To test that it is possible to control material properties of resulting structures, different elements within the 6×6 cathode grid were provided with different voltages in order to create a heart shape structure (Figure 8). Half of the heart shape structure was supplied with 4.7 V at one time, while once the growth time was complete, the other half was supplied with 3.0 V. The intention of this experiment was to determine whether it is possible to indirectly create a specific shape with varying material thickness and what effect varying voltages have on growth rate when using the mineral accretion process.

Figure 8 A heart shape is drawn on the 6×6 grid with the corresponding wire reference to be connected. The heart shape is split into two and supplied with different voltages, each element of the heart is grown for varying time periods. Individual elements are supplied with varying voltages to determine its effects on material composition and properties (see online version for colours)



To examine growth rate of materials under different conditions (e.g. 3 V compared to 4.7 V), one cathode element was disconnected every 3 h for supplied with 3.0 V (Figure 8), and every 2 h for the half supplied in 4.7 V, as shown in Figure 8. The value of 3 h was chosen because initial tests found that it takes 3 h, at 3.0 V to grow enough material to be visible with the human eye. Once all of the elements that were supplied with 3.0 V had finished growing, the wires to be supplied with 4.7 V were connected to grow the other half of the intended heart shape. To determine the effects that varying voltages and currents have on material composition (i.e. percentage amount of limestone compared to other materials present and growth rate), 12 wires were supplied with different voltages, each individual element being supplied with a set voltage for 2 h (Figure 8).

This cathode design should provide a clear heart shape (on the elements supplied with 3.0 and 4.7 V in Figure 8) with different sides of the heart shape growing at different rates and thicknesses. It was expected to see a faster rate of material growth occurring on the half supplied with 4.7 V, with more material aggregation on elements that are supplied with electricity for longer periods of time.

Notably, as discovered in the first experiment, agitation of the seawater solution is important to resist the effects of gravity as materials self-assemble. Consequently, an initial test on the 6×6 grid agitated the solution every 30 min using the aquarium wave maker system; however, from these tests, it was found that agitation of the solution with the grid-shaped cathode structure produced detrimental effects to early growth of crystals, causing grown material to fall off the structure during agitation intervals. To counteract this tendency, this experiment did not use agitation to counteract gravity as material was growing, however, for this experiment, it was believed that the effect of gravity would be less significant because all of the individual elements lie within the same plane (i.e. the grid is flat). Interestingly, it is believed that control of agitation may be a useful parameter for preventing and even reversing material growth in future work.

The experiment ran for a total of 67 h: 27 h for the elements supplied with 3.0 V, 18 h for the elements supplied with 4.7 V and 22 h for the elements supplied with varying voltages. About 300 g of marine salts was dissolved in 6.5 L of water and the 2D cathode grid was then submerged within the solution. Once the first half of the heart shape was grown (3.0 V elements), the cathode was carefully taken out of the solution, the solution was then agitated for 1 min and the cathode grid returned and the other half of the heart shape was grown (4.7 V elements). A new solution was made before carrying out the experiment which supplied elements with varying voltages. Two carbon anodes of radius 6.3 mm and length 75.0 mm were used, because the carbon anodes do not dissolve and contaminate the solution and should not affect the material build-up.

In contrast to the first experiment, which used SEM and XRD methods to perform material analysis, this time X-Ray fluorescence (XRF) was used to analyse the results of this test. XRF was used as it provides a detailed percentage breakdown of the different materials' present which had grown on each of the cathode elements supplied with electrical current. XRF determines the material composition on a small location of the individual cathode elements, as such it may not provide a complete analysis of the whole material composition. XRF analysis was also used as it is much faster than XRD and SEM. To compare growth rates, the radius of material aggregation on each element was measured using a digital Vernier.

4.3 Results

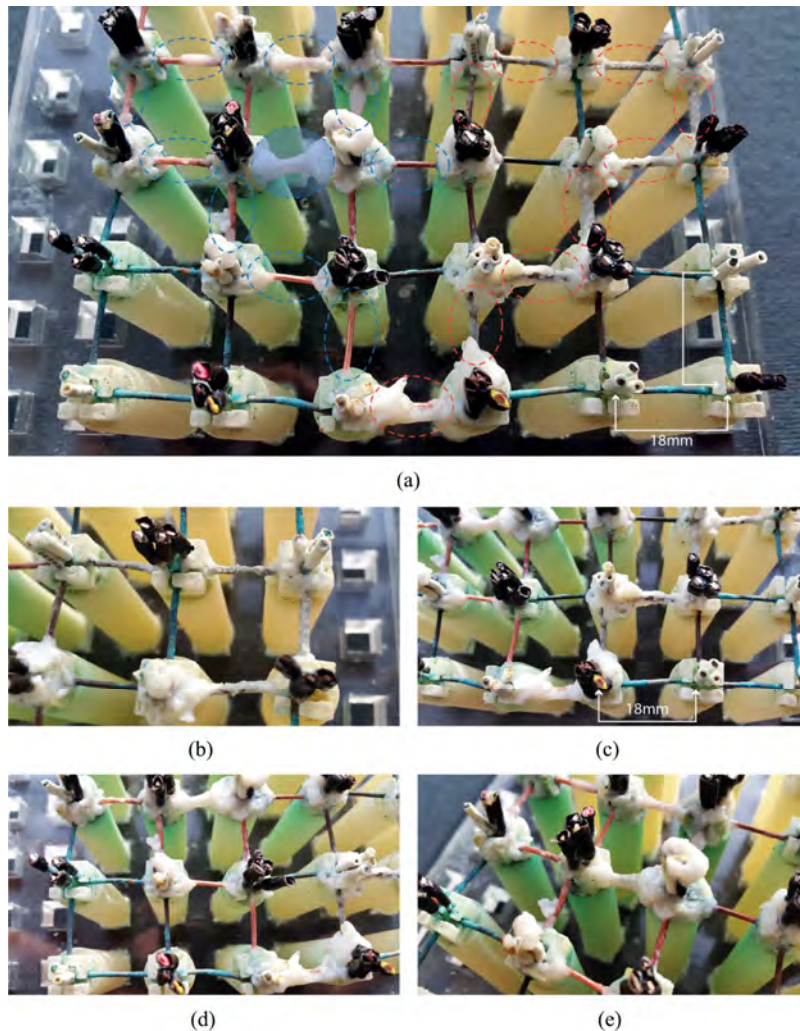
The paper first compares the shape of the grown heart with the intended shape, and then the paper explores the physical properties at different parts of the design.

4.4 Experiment 2: growing a defined shape

Figure 9 shows the completed growth of the heart shape. Growth is shown here by the lighter material, the darker material being a result of the copper oxidising. Examination of the material growth reveals a clear heart shape, which was achieved with the half supplied with 3.0 V, the half supplied with 4.7 V also predominantly grew where it was intended, which completed the heart shape; however, one cathode element (wire 6DB) had unintended growth occurring on it when carrying out the growth of the 4.7 V elements. The unintended growth may be due to the material growth occurring on the intended wire contacting a neighbouring wire and inducing unintended growth. If this is the case, redundancy is required. Introducing redundancy will be able to reverse material growth which occurs on unintended elements. The unintended growth may also be a

result of neighbouring wires contacting one another, this can be addressed by fabricating the scaffolds using AM technology, which affords increased tolerances. The elements supplied with 3.0 V, which were supplied with electrical current for longer time and resulted in increased material build-up, this was also the case for the elements supplied with 4.7 V apart from the last element (wire 6FD). The anomaly may be a result of material continually decaying away from this cathode element during growth, this would suggest the way the materials fabricate themselves is not linear when voltage/turbulence is increased.

Figure 9 (a) The final heart shape growth reveals material growth location and amount can be controlled. (b) and (c) reveal varying surface textures and varying growth location over time when supplied with 3.0 V. (d) and (e) reveal a smooth surface texture, which was produced with initial growth predominating at the bends which became more uniform over time when supplied with 4.7 V (see online version for colours)



4.5 Experiment 2: localised material properties

The primary objective was to grow a predefined heart shape on the 2D grid that had varying material volumes. The heart shape was successfully grown. Additionally, various material properties appeared on the individual elements of the heart shape and also the elements supplied with varying voltages over time, this highlights that both time and environmental manipulations can result in increased control over material properties. The findings are revealed from examination during material growth phase and the resultant structures from material growth (Figures 9–13).

Figure 10 This reveals the growth volume and rate increases for both 3.0 and 4.7 V longer the cathode element is supplied with electrical current. Error bars highlight the range between average growth and minimum and maximum growth at point on each cathode wire (see online version for colours)

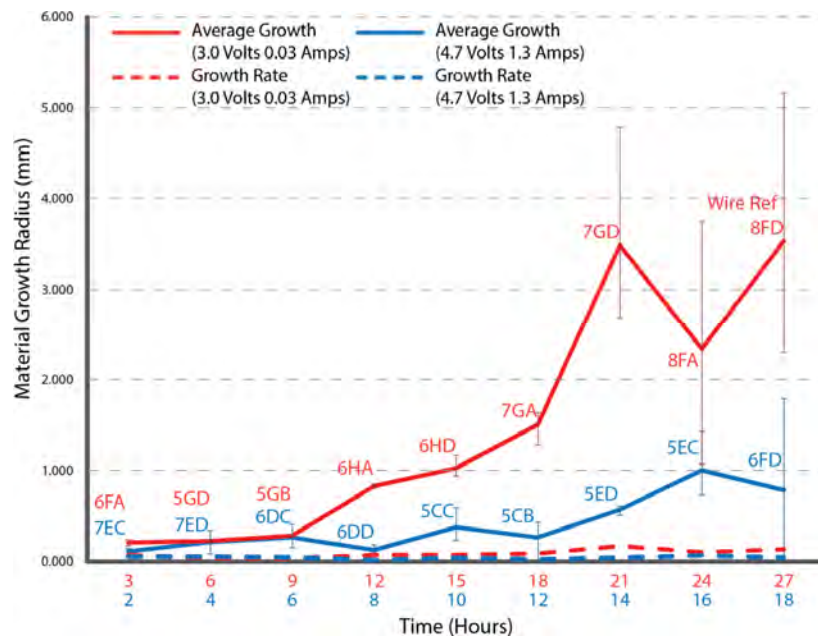


Figure 11 This reveals an initial trend that material growth amount and rate increase as voltage/current increases but cannot be sustained due to the materials' increasing fragility (see online version for colours)

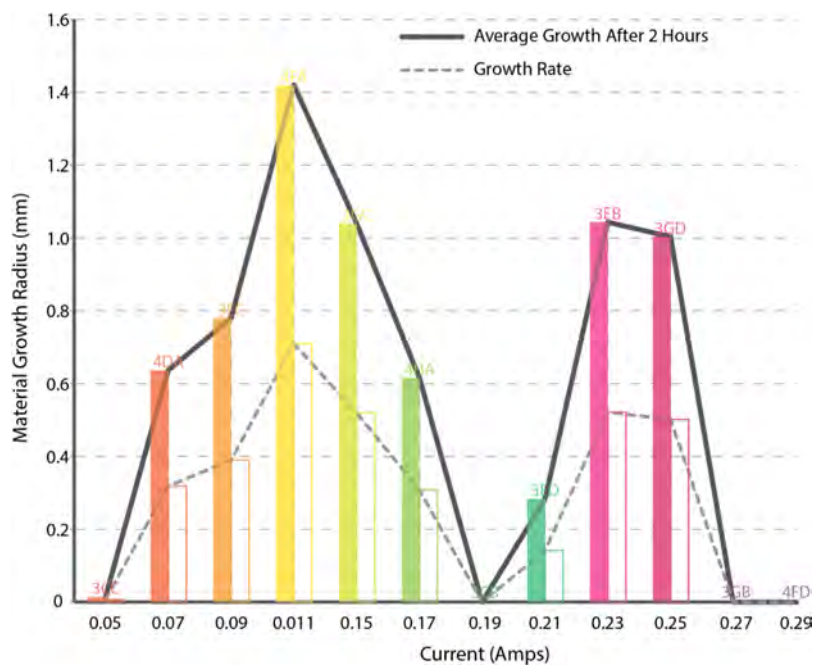


Figure 12 XRF analysis of the elements supplied with varying voltages reveals that the material composition can be tuned by varying voltages. An increase in voltage results in an increase in chlorine with the reduction of all other materials (see online version for colours)

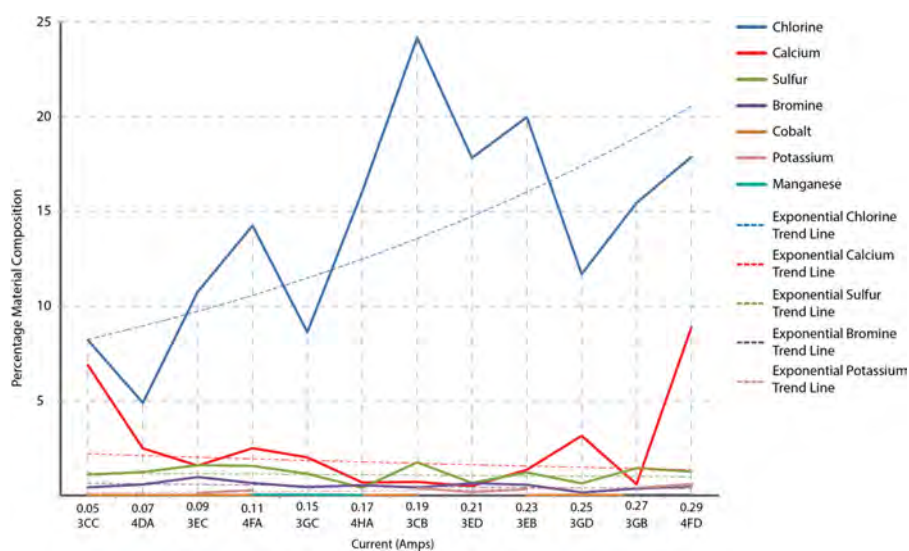
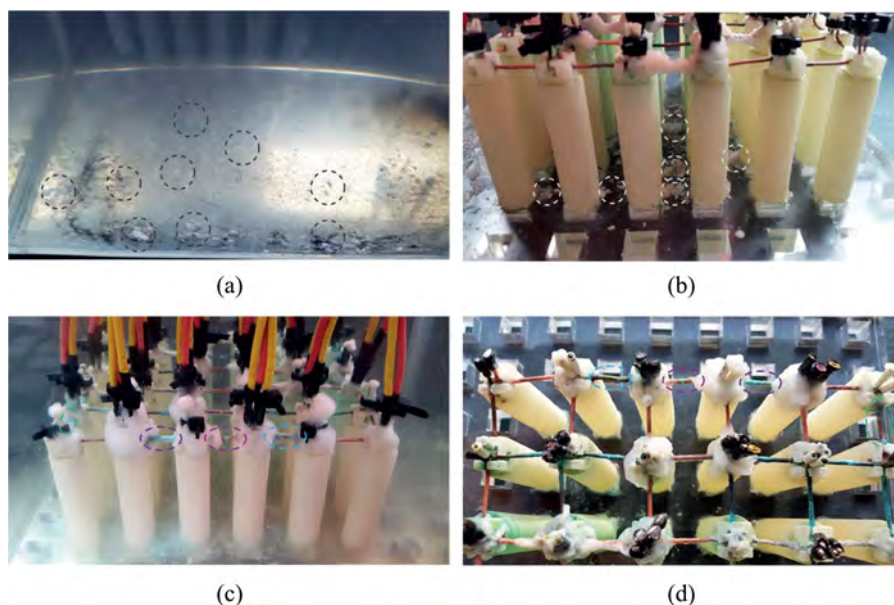


Figure 13 (a) and (b) Material growth would decay away from the cathode elements as it grew. (c) and (d) Material decay became more apparent as the voltages increased (see online version for colours)



Figures 10 and 11 shows plots of material deposition over time. Figure 10 reveals that material deposition increased for both voltage values (3.0 and 4.7 V), respectively, the longer they were supplied with electrical current, this establishes control over the amount of material grown at a specific location. Interestingly more material volume and deposition occurred with the elements supplied with 3.0 V, which were not the expected result due to the lower voltage, this suggests that introducing more turbulence during early stages of material deposition has a detrimental effect. Increased turbulence in this case was a result of increasing voltages, this resulted in an increased amount of hydrogen being produced at the cathodes which appears to have prevented material deposition. Interestingly, the growth rates for both voltages (3.0 and 4.7 V) are quite similar, which both increase over time.

The ability to grow increasing amounts of various materials at increasing rates in specific locations enables the structures to adapt at a faster rate regarding material location. Figure 11 shows an initial trend of increasing material growth amount corresponds with increasing voltages between 3.6 and 4.5 V (0.05–0.11 A), this then falls off. Comparing growth rate (Figure 11) with the material composition (Figure 12) produced from varying voltages reveals that most material deposition occurred if the presence of chlorine remained below 15%. As chlorine rose over a threshold of 15%, the material became more fragile, this was determined by reduced or no material deposition being recorded in these samples. Figure 12 reveals XRF analysis of the cathode elements supplied with varying voltages, the analysis highlights that material composition and functionality can be tuned by manipulating the environment (voltage).

The paper now discusses the four material properties and behaviours observed during this experiment.

First, *varying surface textures* on the heart shape elements were supplied with 3.0 V. A smoother texture was produced on elements supplied with electrical current for longer periods of time, this is compared to an initially granular texture produced from shorter periods of time. A uniform surface texture was created on the heart shape supplied with 4.7 V (Figure 9). Control of surface textures can be tuned to increase surface area, which could result in surface ornamentation of architectural structures.

Second, *uniform material growth* on the cathode elements was achieved on the 3.0 V elements that were supplied with electrical current for shorter periods of time (up to 15 h). On these elements supplied with electrical current for longer periods, material build-up became more predominant at the bends of the cathode elements. *Non-uniform material growth* on the 4.7 V elements was created as the majority of the material build-up occurred at the bends of the cathode elements. Interestingly, this allows an increase in control over localised material properties to emerge on individual cathode elements. Initial smooth surface textures can be grown at specific bends/locations and then more granular materials created between these materials, this incorporates both time and environmental manipulations which can govern material properties.

Third, *material decay* was observed during and after material growth (Figure 13), this was most significant as the voltages increased, at this point, an increasing amount of material would deteriorate away from the cathode elements and build-up on the base of the jig and tank. The ability to reverse or remove material from cathode elements introduces redundancy within the system, this maintains control of material growth location if material growth produced on one element contacts a neighbouring element and initiates growth.

Finally, *varying mechanical strengths* and compositions were achieved by varying voltages supplied to the cathode elements, this was determined by XRF analysis (Figure 12). As voltage increased, the amount of calcium present in the material was reduced and resulting in an increasingly fragile material. Elements supplied with more voltage appeared very fragile upon examination (Figure 13), this was revealed as the material deteriorated once it was taken out of the tank as a result of the solutions' surface tension, this establishes that varying material qualities and functionalities can be achieved by altering the environment (voltage) as materials grow.

5 Conclusion

This research represents initial steps towards an ultimate goal of creating truly adaptive physical structures, demonstrated by the results using the mineral accretion process. XRD analysis of the fence cathode demonstrates that it is possible to create a multi-material system with this approach (Figures 3 and 6). The SEM analysis of the two individual wire cathodes shows that varying electrical current controls the material type deposited (Figures 4 and 5). Finally, the 6×6 grid study demonstrates control over material placement and composition by varying electrical current to modify crystal growth. Significantly, these results obtained establish that it is possible to grow structures from an abundant base material (seawater), which can tune and adapt multiple material properties (shape, composition, volume, location, rate, texture) by imposing external stimulus (voltage, pH, solution agitation) upon the environment in which they grow.

By growing structures through aggregation two major benefits are achieved. First the structures can be grown within a 3D matrix scaffold which can change shape and material

properties globally or locally. Second the fabrication process is scalable as the material deposited on the scaffolds is based on the material scale, this is because the molecules that make up the deposited material self-assemble.

Further work could explore the use of design simulations to govern environmental manipulations (voltage amount, voltage location and gradient electrical currents) through hardware to automate the fabrication process. Automating the fabrication process may enable

- 1 Logic from digital designs to be instilled within the fabrication process and resultant physical structures.
- 2 Adaptive, flexible and tuneable capacities of design simulations to be physically instilled within the structures (i.e. structures fabricated within the constraints of a 3D cathode matrix could change shape and material properties).
- 3 Increasingly, complex structures could be fabricated by imposing multiple design demands, which results in integrated structures (Wiscombe, 2010, 2012) and multi-functional materials (Oxman, 2012) being created. Notably, this paper shows proof of concept that material properties can be tuned (material volume, surface texture, composition) over time through environmental manipulations (voltage), and that this enables functionally graded materials and structures (Richards and Amos, 2014a, 2014b) to be fabricated.
- 4 A continual discourse between design, fabrication and materials could be achieved in real time.

Acknowledgements

Thank you to Nathan Halcovitch and Sara Baldock for carrying out the material analysis. This work is funded by the Digital Economy program (RCUK Grant EP/G037582/1), which supports the HighWire Centre for Doctoral Training (<http://highwire.lancs.ac.uk>).

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